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Effect of Co-Doping on the Photocatalytic, Anti-Bacterial and Hydrogen Evolution Properties of Ag-TiO_2 Nanoparticles

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ABSTRACT

Silver-doped titanium dioxide (Ag-TiO_2) nanoparticles have emerged as a promising material with implementation in photocatalytic water treatment, antibacterial applications, coatings, and solar-assisted hydrogen evolution. It has become a subject of extensive research because of its ease of production and overall stability. However, there are certain challenges. Practical application of Ag-TiO_2 nanoparticles is limited because of their underwhelming photocatalytic activity under visible light, silver nanoparticle leaching, and potential for environmental toxicity. Recent developments in the co-doping of Ag-TiO_2 with elements such as carbon, nitrogen, fluorine, copper, yttrium, zinc oxide, strontium, and zirconium have shown significant improvements in photocatalytic activity, antibacterial efficacy, and hydrogen evolution. Doping of certain elements and modifying the surface morphology have enhanced the overall performance by increasing surface area, reducing electron-hole recombination, and adjusting the band gap energy. This review assembles recent developments in Ag-TiO_2 co-doping, featuring its promising applications and also highlighting the requirement for further studies to ensure effective execution.

Keywords: Photocatalytic Activity, Ag-TiO_2, Anti-Microbial, Hydrogen Evolution, Co-Doping.

1. Introduction

Human civilization is facing multiple challenges, such as health, natural resources, and energy demands, as the population races towards 8 billion. Water is one of the most valuable natural resources of our planet. This may seem counterintuitive, given that approximately 75% of the planet's surface is covered by water and a huge amount of water is stored in glaciers and underground aquifers, but it is justifiable, as only 2.5% of this water is actually usable and accessible due to low freshwater content, geographical restrictions, logistical issues, and geopolitics. Due to industrialization, rapid urbanization, and climate change, water scarcity has become a major problem in some parts of the world. Even in a country like Bangladesh, with roughly 700 rivers, there is a shortage of clean, drinkable, and usable water. According to the latest report from WaterAid, only 59.11% of the total population has access to safely managed water, 14.94% of the total population is under a piped water supply, and about 86% of the poorest households have been proven to use E.coli-contaminated water [1]. The "Global Commission on the Economics of Water" reported that about 40% of the total population of our planet will experience water shortage by 2030 [2]. In the capital city of Dhaka, about 87% of the water supply comes from groundwater/deep aquifers [3]. This causes the water level to drop by 2-3 meters yearly [4]. Due to increased water usage, authorities are now drawing water from a deep aquifer thousands of feet underground [5]. Groundwater usage in such large quantities can be detrimental, creating sinkholes and reducing flow to nearby rivers. Only 2 sewage water treatment plants are operational, but with reduced capacity. Also, there is no centralized industrial wastewater management system available in any city in Bangladesh. Hence, it has become evident that water treatment is required for a sustainable water supply. Considering the economic constraints of Bangladesh, a solution is needed that can be deployed quickly, cost-effectively, and with little to no adverse environmental effect. The recent outbreak of COVID-19 made us more aware of disease transmission through contact. SARS-CoV-2, the virus responsible for COVID-19, can survive on surfaces for up to three days, depending on the material [6]. This greatly increased the rate of transmission, as surfaces that appeared clean to the naked eye became potential carriers. Similarly, many disease-causing bacteria can grow and spread through surface contact. Over time, increasingly potent pathogens are being discovered that can cause mass exterminations, which can severely impact human civilization and may take years or decades to recover. Filling the energy demand for 8 billion people is another massive challenge. Report of Energy Institute's Statistical Review of World Energy 2025, 74th edition states that about 86.57% of the planet's total energy demand was fulfilled using fossil fuels in 2024, which contributed to about 35,491.8 million tons of CO_2 released in the atmosphere with an annual growth rate of 1.1% over 2023 [7]. In one year, between 2023 and 2024, the average temperature of the planet increased by about 0.12 degrees Celsius. According to data from Copernicus Climate Change Service, global temperature was above the Paris Agreement-suggested threshold of 1.5 degrees Celsius above the pre-industrialization level for 11 to 12 months consecutively, with the highest temperature recorded back-to-back in previous years [8]. Although alternative energy sources have been developed at an increasing rate over the past few years,

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none of them are actually carbon neutral, with the most efficient being nuclear energy.

From a materials scientist's perspective, Ag-TiO_2 (Ag-doped TiO_2) nanoparticles can be a promising solution. Ag-TiO_2 has demonstrated enhanced photocatalytic activity, resulting in the degradation of dyes and organic compounds, an antimicrobial effect, and solar-assisted hydrogen evolution. It acts like a catalyst and hence can be used several times without loss in efficacy. Due to reusability, it can be cost-effective as well.

In spite of all these positive aspects, there are some drawbacks, mainly due to the possibility of Ag leaching and intoxicating the environment, low performance under visible light due to lower Ag concentration, reduced efficacy in case of Ag particle agglomeration on the NP surface, etc. Co-doping Ag-TiO_2 with other nanoparticles can alleviate these limitations to some extent while retaining the original properties. Recent works done in this matter exhibit higher photocatalytic ability, increased efficiency in dye degradation, and overall stability.

2. Fundamentals of Ag-Tio_2 Nanoparticles

Ag-TiO_2 nanoparticles are produced by doping Ag into TiO_2. This process is done by various methods such as sol-gel, hydrothermal, green synthesis, etc. TiO_2 can be found in various forms such as rutile, anatase, brookite, etc. Anatase phase is the most effective one in case of photocatalysis due to higher reactive oxygen species generation, slower electron hole recombination, higher surface area, fewer defect sites, etc. Ag is doped with TiO_2 mainly to reduce the band gap of anatase TiO_2, which typically has a band gap of 3.2eV. This band gap of anatase TiO_2 corresponds to absorption of UV light only. With a wavelength of about 387 nm. Doping Ag reduces this band gap to a varying degree, depending on the Ag content [9], and shifts the absorption range into the visible light zone. Visible light photocatalysis is much easier than UV due to its availability. Ag also induces a plasmonic effect, which is responsible for the increased photocatalytic activity [10].

The basic working process of Ag-TiO_2 is to create ROS or Reactive Oxygen Species. Reactive Oxygen Species are highly active molecules that contain oxygen. Some notable examples are H_2O_2, $O_2{\cdot}^-$, $\cdot OH$, $^1O_2$ etc. These molecules can cause oxidation of organic molecules, proteins, lipids, and DNA. ROS is the cornerstone of Ag-TiO_2's ability to treat water, inhibit bacterial growth, and produce H_2 by splitting H_2O using light.

3. Challenges of Ag-Tio_2 Nanoparticles

Major drawbacks of Ag-TiO_2 nanoparticles are the possibility of Ag leaching, low photocatalytic activity under visible light, narrow allowable dopant concentration [11], and the chance of reduced efficacy due to an increase in agglomeration [12]. Leaching can cause lower efficacy as the Ag concentration is reduced, resulting in a lower effective surface area, reduced stability, and reusability, and raises environmental concern as Ag can be hazardous to certain life forms, including humans. Ag-TiO_2 nanoparticles have been found to be detrimental to human respiratory health [13], [14], [15], [16]. Ag is also one of the most precious metals, which makes the production cost relatively higher. Silver leaching from Ag-TiO_2 nanoparticles typically ranges from less than 0.4% of total silver content in pure water over 36 hours to up to 4% in saline solution within one week, with leaching rates highly dependent on water chemistry, time, and nanoparticle morphology; under specific conditions, Ag ion release can reach concentrations such as 0.4 ppm (1 hour), 0.26 ppm (24 hours), and 0.005 ppm (7 days) for films immersed in water [17], [18], [19], [19], [20]. Regarding cytotoxicity, human mesenchymal stem cells exhibit cytotoxic effects at a silver concentration of 12.5 μg/mL silver ions (Ag^+) are acutely toxic to microorganisms, flora, and fauna in aquatic environments, even in low concentrations of (1-5μg/L), leading to mortality, morphological defects, and physiological disturbances. Empirical studies indicate that the very sensitive taxa, such as the cladoceran *Daphnia magna*, and other species of fish develop immobilization and abnormal development after being exposed to Ag^+ [21].

4. Co-Doping Strategies

The purpose of co-doping is to enhance the performance of the existing nanoparticle technology and to resolve the drawbacks. The target is to increase surface area, increase stability, reduce band gap, and cost of production. To achieve these objectives, an appropriate material should be identified. Investigations have found that noble metals are the most effective in achieving higher photocatalytic activity when used as dopant elements [22].

Doped TiO_2s have several electronic interaction mechanisms, where only one or more of these mechanisms can be used at the same time, which is what makes doped TiO_2 systems unique in terms of controlling TiO_2 properties.

One of the basic processes is the formation of mid-gap states, which are specific to transition metals, including Cu and Fe, and non-metals, including N. Dopant atomic orbitals (e.g., Cu 3d, N 2p) interact weakly with Ti 3d and O 2p orbitals, to form discrete impurity states within the TiO_2 band gap. These localized states offer intermediate energy levels to electron excitation between the valence band and the conduction band and thus reduce the amount of photon energy needed. This mechanism, therefore, extends the light absorption of TiO_2 to the visible spectrum and explains the narrowing of the band gap in relation to substitutional or interstitial doping [23].

The other important process is the charge transfer and electron trapping, which is mainly through metallic dopants, e.g., Ag. When Ag nanoparticles are deposited on TiO_2 surfaces, the nanoparticles create Schottky barriers (the difference between the Fermi levels) between the surfaces, resulting in the flow of photogenerated electrons from TiO_2 to Ag, which acts as a sink of electrons. This trapping increases the lifetime of the electrons by inhibiting recombination with holes, hence improving the efficiency of the photocatalyst. Unlike mid-gap formation of states, this process alters the dynamics of charge-carriers both by energetically-favored interfacial alignment and not by the formation of new electronic levels [24].

Oxygen vacancies and defect states are common during doping, particularly during the replacement of Ti^{4+} by low-valence dopants (e.g., Cu^{2+}) or the replacement of oxygen with non-metals (e.g., N). These vices create Ti^{3+} centers with localized electrons in the conduction band, which create shallow levels of donors that support electron flow but can also cause unwanted recombination at elevated concentrations. The vacancies, therefore, have a role in modulating photocatalytic activity and reactions to offset beneficial charge-transfer reactions in connection with the dangers of recombination.

The band-edge shifts take place through orbital hybridization and lattice distortion, which are caused by dopants. Dopants change the overlap and the energies of Ti 3d and O 2p orbitals to shift the valence or conduction band edges. As an example, testable examples include the effect of substitutional nitrogen on the highest, most energetic band in the spectrum, which now has its 2p orbital at a higher energy level than the oxygen orbital, and the testable effect of strontium doping on crystal structure, where the crystal field is altered, and therefore the band edges are indirectly shifted. These variations fine-tune the band gap and also modify the carrier mobility and light absorption cutoffs.

Lastly, some dopants (e.g., Li^{+}, Fe^{2+}/ Fe^{3+}) saturate intrinsic trap states and hence, decrease deep recombination centers. This passivation improves charge transport and photocurrent by reducing electron-to-hole recombination through the elimination of the trap states by neutralization [25].

All of these processes, including mid-gap state formation, electron trapping, vacancy-associated defect states, band-edge shifts, and trap passivation, determine the ability of dopants to tune the electronic structure of TiO_2 and photocatalysis. Their importance is dependent on the chemical nature of the dopant, the site of location (substitutional or interstitial), and the concentration of the dopant in the lattice, which allows the precise design of TiO_2 materials to be applied in varied applications.

5. Effect of Co-Doping on the Properties of Ag-TiO_2

Recent studies have explored co-doping Ag-TiO_2 with various elements to overcome the limitations of Ag-TiO_2. These studies show significant improvement in photocatalytic ability, anti-microbial effect, band gap modulation, hydrogen evolution prospect, and are known to have very little toxicity when in ionic forms or bound to the TiO_2 lattice. The problem with Ag^{+} can hence be minimized by reducing silver content and substituting Ag with other elements such as $Zr^{4+,}$ which creates substitutional doping in TiO_2 crystal and doesn't leach due to strong atomic bonding, unlike silver, which only occupies interstitial sites. To provide a structured analysis, the discussion is divided into subsections addressing photocatalytic, antimicrobial, band gap modulation, and hydrogen evolution properties. In each subsection, recent and most widely available data have been tabulated. Note that values provided here are strictly collected from literature and are not standardized (re-analyzed). This is due to different work processes, dopant/co-dopant concentration, testing conditions, etc. Values mentioned here demonstrate the highest experimental performances for each dopant/co-dopant and give an insight into co-dopant selection for further studies.

5.1 Photocatalytic Activity

Photocatalytic activity is the most important measurement of the performance of co-doped Ag-$TiO_{2,}$ as it is the driving force for water treatment, anti-microbial effect, and hydrogen evolution. We can get an idea of its photocatalytic activity by measuring the degradation of sample compounds. Higher results indicate better performance. The table summarizes and compares the effect of co-doping on the photocatalytic activity of co-doped Ag-TiO_2.

Table 1 Photocatalytic Degradation Efficiency of Co-doped Ag-TiO_2 Samples

Co-dopant	Target compound	Performance	References
Nitrogen	Methyl Blue	98.82% degradation in 6 hours	[26]
Carbon and Fluoride	Rhodamine B	85% dye degradation in 4 hours	[27]
Yttrium	Methyl Blue Methyl Red Methyl Orange	A 99% MB, 56% MR, and 63% MO degradation over 12 hours.	[28]
SiO_2	Methyl Blue	63.3% after 10 hours	[29]
Copper	Red Bengal Dye	81.07% in 1 hour	[30]
Zirconium	Methyl Blue	64.2-92.8 in 90 min	[31]
SiO_2	Methyl Blue	63.3% after 10 hours	[29]

5.2 Anti-Microbial Effect

ROS generated by the photocatalytic effect significantly degrades organic compounds such as lipids, proteins, DNA, etc. ROS attacks the cell membranes, DNA, and RNA of microbes and destroys them. This results in disinfection and inhibition of further microbial growth. The table summarizes and compares the effect of co-doping on the photocatalytic activity of co-doped Ag-TiO_2.

Table 2 Anti-Microbial Effect of Co-doped Ag-TiO_2 Samples

Co-dopant	Target microbe	Performance	References
Strontium	*E. coli*	96% inhibition when incubating for 5 hours	[32]
SiO_2	*E. coli*	98.4% reduction in 1 hour	[29]
Copper	*E. coli* *S. aureus* *A. niger*	75-99.7% reduction in 5-10 days	[33]
ZnO	*E. coli* *B. subtilis* *P. aeruginosa* *S. aureus*	Strong inhibition activity with 15-32 µg/ml concentration	[34]

5.3 Hydrogen Evolution

ROS reacts with the water molecules, splitting them into hydrogen and oxygen molecules. Hydrogen molecules can be collected and, through processing, can be used for power generation by ignition or fuel cell operation. The table summarizes and compares the effect of co-doping on the hydrogen evolution performance of co-doped Ag-TiO_2.

Table 3 Co-doping-induced Hydrogen Evolution

Co-dopant	Performance	References
Strontium	64.3 mmol h^{-1}	[32]
Copper	17.1 μmol h^{-1} g^{-1}	[30]

5.4 Band Gap Modulation

Effective photocatalytic activity under visible light heavily depends on the band gap of the nanoparticle compound. Band gap narrowing enhances photocatalytic activity by allowing excitation of electrons with lower energy photons, thus enabling visible-light-driven electron-hole pair generation. The presence of dopant-induced states inside the band gap or shifts in the valence and conduction band edges facilitates these transitions. Consequently, electrons can be excited by visible light to these intermediate states or into the conduction band more efficiently, leading to increased generation of reactive species such as hydroxyl radicals or superoxide anions critical for photocatalysis. For usage under visible light suitable band gap range should be between 3.1eV to 1.77 eV, which corresponds to a 400nm-700nm range of wavelengths. Typical anatase TiO_2 band gap is about 3.2eV, which is why it absorbs mostly UV light. The table shows the band gap resulting from the co-doping of different elements into/with Ag-TiO_2.

Table 4 Effect on TiO_2 bandgap after doping.

Co-dopant	Resulting Band-gap	References
Nitrogen	1.5-1.6eV	[26]
Carbon and Fluoride	2.56eV	[27]
Yttrium	3.42-3.51eV	[28]
Copper	2.86eV	[30]
Zirconium	2.66eV	[31]

5.5 Miscellaneous

Recent studies done on the topic of co-doped Ag-TiO_2 have also provided some unexpected results. Results include removal of gas phase NH_3 using N co-doped Ag-TiO_2 [26], reduced electron hole recombination by La co-doped Ag-TiO_2 [35], higher conversion efficiency using 3% Cu doped Ag-TiO_2 when used in dye sensitized solar cells [36], and TiO_2 crystallite size reduction when doping with Zr and Ag at an increased amount[31]. Further studies regarding these can widen the usage of co-doped Ag-TiO_2 nanoparticles.

6. Limitations and Research Gaps

Recent studies have shown promising results for photocatalytic activity, band gap modulation, antimicrobial effects, and hydrogen evolution. However, several important research gaps remain. For practical implementation in water treatment, testing against major household, agricultural, and industrial water pollutants must be done, which include types of organic compounds, pesticides, herbicides, pharmaceuticals, and heavy metal compounds. Some major limitations regarding both household and industrial applications are the recuperation of nanoparticles to be reused and the need for light energy. Recuperation of nanoparticles can be done via the reverse osmosis method. In terms of antimicrobial activity, nanoparticles should be tested against various types of contagious bacteria, viruses, and other pathogens. For hydrogen evolution prospects, in-depth studies are required in order to find the best doping combination and strategy. A major limitation regarding this is that at the current level, the hydrogen production rate is very limited. Hydrogen evolution also produces oxygen at the same time, which is beneficial as oxygen can be used for medical purposes, as an oxidizing agent, or to increase fuel efficiency by mixing oxygen into the fuel mix, but it poses a challenge as separating hydrogen from oxygen gas mix needs additional equipment for membrane separation. Efficient implementation design, including scalability, cost, and or potential implementation, must be studied and trialed to ensure sustainable applications.

7. Conclusion

Ag-TiO_2 co-doped with other elements shows an increase in photocatalytic activity, exhibits improved antimicrobial properties, effective band gap reduction, and also shows promising results for hydrogen evolution. The performance of doped nanoparticles is greatly affected by dopant concentration, size of nanoparticles, surface area, and morphology. However, certain concerns, such as the leaching of Ag ions, stability of the nanoparticles, and environmental and biological impacts of doped nanoparticles, have been under-addressed. Future studies should investigate the optimal dopant concentrations and combinations, a cost-and energy-efficient approach in terms of synthesis, long-term stability, methods of application such as filtration devices or anti-microbial coatings, and evaluate application against several major water pollutants and clinically relevant pathogens, as well as up-scaled strategies towards sustainable applications in water purification, antimicrobial defense, and clean energy production.

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